

PCT/PTO 06 FEB 2002

TRANSMITTAL LETTER TO THE UNITED STATES  
DESIGNATED/ELECTED OFFICE (DO/EO/US)  
CONCERNING A FILING UNDER 35 U.S.C. 371

ATTORNEY'S DOCKET NUMBER

1888

U.S. APPLICATION NO. (If known, see 37 CFR 1.5)

10/049361

INTERNATIONAL APPLICATION NO.

PCT/US00/21874

INTERNATIONAL FILING DATE

10 August 2000 (10.08.00)

PRIORITY DATE CLAIMED

10 August 1999 (10.08.99)

TITLE OF INVENTION

COSMETICS

APPLICANT(S) FOR DO/EO/US

NATIONAL STARCH AND CHEMICAL INVESTMENT HOLDING CORPORATION

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. ☒ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
2. ☐ This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.
3. ☐ This is an express request to begin national examination procedures (35 U.S.C. 371(f)). The submission must include items (5), (6), (9) and (21) indicated below.
4. ☐ The US has been elected by the expiration of 19 months from the priority date (Article 31).
5. ☒ A copy of the International Application as filed (35 U.S.C. 371(c)(2))
  - a. ☐ is attached hereto (required only if not communicated by the International Bureau).
  - b. ☒ has been communicated by the International Bureau.
  - c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US).
6. ☐ An English language translation of the International Application as filed (35 U.S.C. 371(c)(2)).
  - a. ☐ is attached hereto.
  - b. ☐ has been previously submitted under 35 U.S.C. 154(d)(4).
7. ☒ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))
  - a. ☐ are attached hereto (required only if not communicated by the International Bureau).
  - b. ☐ have been communicated by the International Bureau.
  - c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
  - d. ☒ have not been made and will not be made.
8. ☐ An English language translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
9. ☒ An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).
10. ☐ An English language translation of the annexes of the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).

## Items 11 to 20 below concern document(s) or information included:

11. ☐ An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
12. ☒ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
13. ☒ A FIRST preliminary amendment.
14. ☐ A SECOND or SUBSEQUENT preliminary amendment.
15. ☐ A substitute specification.
16. ☐ A change of power of attorney and/or address letter.
17. ☐ A computer-readable form of the sequence listing in accordance with PCT Rule 13ter.2 and 35 U.S.C. 1.821 - 1.825.
18. ☐ A second copy of the published international application under 35 U.S.C. 154(d)(4).
19. ☐ A second copy of the English language translation of the international application under 35 U.S.C. 154(d)(4).
20. ☐ Other items or information:

US APPLICATION NO. (If known, 37 CFR 1.51) <b>10/049361</b>		INTERNATIONAL APPLICATION NO. <b>PCT/US00/21874</b>		ATTORNEY'S DOCKET NUMBER <b>1888</b>	
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21. <input checked="" type="checkbox"/> The following fees are submitted: <b>BASIC NATIONAL FEE (37 CFR 1.492 (a) (1) - (5)):</b> Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO. .... <b>\$1040.00</b>  International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO ..... <b>\$890.00</b>  International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO ..... <b>\$740.00</b>  International preliminary examination fee (37 CFR 1.482) paid to USPTO but all claims did not satisfy provisions of PCT Article 33(1)-(4) ..... <b>\$710.00</b> International preliminary examination fee (37 CFR 1.482) paid to USPTO and all claims satisfied provisions of PCT Article 33(1)-(4) ..... <b>\$100.00</b> <b>ENTER APPROPRIATE BASIC FEE AMOUNT =</b>				<b>CALCULATIONS PTO USE ONLY</b>	
				\$ 890.00	
Surcharge of <b>\$130.00</b> for furnishing the oath or declaration later than <input type="checkbox"/> 20 <input checked="" type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(e)).				\$ 130.00	
CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE		
Total claims	14 - 20 =	0	x \$18.00	\$ 0	
Independent claims	1 - 3 =	0	x \$84.00	\$ 0	
MULTIPLE DEPENDENT CLAIM(S) (if applicable)				+ \$280.00	
<b>TOTAL OF ABOVE CALCULATIONS =</b>				<b>\$ 1,020.00</b>	
<input type="checkbox"/> Applicant claims small entity status. See 37 CFR 1.27. The fees indicated above are reduced by 1/2.				+	
<b>SUBTOTAL =</b>				<b>\$ 1,020.00</b>	
Processing fee of <b>\$130.00</b> for furnishing the English translation later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(f)).				\$	
<b>TOTAL NATIONAL FEE =</b>				<b>\$ 1,020.00</b>	
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). <b>\$40.00</b> per property +				\$ 40.00	
<b>TOTAL FEES ENCLOSED =</b>				<b>\$ 1,060.00</b>	
				Amount to be refunded: \$	
				charged: \$	

a. ☐ A check in the amount of \$ \_\_\_\_\_ to cover the above fees is enclosed.

b. ☒ Please charge my Deposit Account No. 14-0455 in the amount of **\$1,060.00** to cover the above fees.  
 A duplicate copy of this sheet is enclosed.

c. ☒ The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any  
 overpayment to Deposit Account No. 14-0455. A duplicate copy of this sheet is enclosed.

d. ☐ Fees are to be charged to a credit card. **WARNING:** Information on this form may become public. **Credit card  
 information should not be included on this form.** Provide credit card information and authorization on PTO-2038.

**NOTE:** Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR  
 1.137 (a) or (b)) must be filed and granted to restore the application to pending status.

SEND ALL CORRESPONDENCE TO

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 NAME  
 33,506  
 REGISTRATION NUMBER

CASE 1888

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

IN RE APPLICATION OF

TSUZUKI, ET AL.

S.N.

FILED: CONCURRENTLY HERewith

FOR: COSMETICS

Group Art Unit:

Examiner:

Commissioner of Patents and Trademarks

Washington, D.C. 20231

PRELIMINARY AMENDMENT

Sir:

In the above-identified application, Applicants respectfully request the following preliminary amendment be entered and the claims considered in light thereof.





STATUS OF THE CLAIMS

Claims 1-13 were filed as PCT/US00/21874 internationally filed 10 August, 2000.

Claims 1-13 have been cancelled.

Claims 14-27 have been added. A marked up copy of the claims as amended is attached as Appendix A.

Claims 14-27 are presented for consideration. A clean copy of the claims as currently pending is attached as Appendix B.

REMARKS

Claims 1-13 have been cancelled and new claims 14-27 have been added. Claims 14-27 have descriptive basis in the claims as filed in PCT/US00/21874, but have been amended to conform to standard US patent practice.

In view of the foregoing, Applicant respectfully requests early action in this application.

Respectfully submitted,



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Dated: 30 Jan 2002

Appendix A  
(marked up copy of amended claims)

- [
1. A cosmetic comprising an amphoteric urethane resin having carboxyl group(s) and tertiary amino group(s) in a molecule, and silicone polymer.
  2. The cosmetic according to claim 1, wherein said silicone polymer is a nonionic silicone polymer.
  3. The cosmetic according to claim 1, wherein said silicone polymer is a polyether-modified silicone polymer.
  4. The cosmetic according to claim 1, wherein said silicone polymer is a phenyl-modified silicone polymer.
  5. The cosmetic according to claim 1, wherein said silicone polymer is an amino-modified silicone polymer.
  6. The cosmetic according to claim 1, wherein said silicone polymer is an alkyl-modified silicone polymer.
  7. The cosmetic according to claim 1, wherein said silicone polymer is an alkoxy-modified silicone polymer.
  8. The cosmetic according to claim 1, wherein said silicone polymer is a cyclic silicone polymer.
  9. The cosmetic according to any one of claims 1 to 8, wherein said silicone polymer is in the form of an aqueous solution or a water dispersion.
  10. The cosmetic according to any one of claims 1 to 9, wherein said amphoteric urethane resin has structural unit(s) derived from ethylene oxide in its structure.
  11. The cosmetic according to any one of claims 1 to 9, wherein said amphoteric urethane resin has polysiloxane bond(s) in its structure.
  12. The cosmetic according to any one of claims 1 to 11, wherein said amphoteric urethane resin is in the form of an aqueous solution.
  13. The cosmetic according to any one of claims 1 to 12, which is at least one selected from the group consisting of hair cosmetics, skin care cosmetics and make-up cosmetics.]
  14. A cosmetic comprising an amphoteric urethane resin having at least one carboxyl group and at least one tertiary amino group in a molecule, and a silicone polymer.
  15. The cosmetic according to claim 14, wherein said silicone polymer is a nonionic silicone polymer.
  16. The cosmetic according to claim 14, wherein said silicone polymer is a polyether-modified silicone polymer.
  17. The cosmetic according to claim 14, wherein said silicone polymer is a phenyl-modified silicone polymer.

18. The cosmetic according to claim 14, wherein said silicone polymer is an amino-modified silicone polymer.

19. The cosmetic according to claim 14, wherein said silicone polymer is an alkyl-modified silicone polymer.

20. The cosmetic according to claim 14, wherein said silicone polymer is an alkoxy-modified silicone polymer.

21. The cosmetic according to claim 14, wherein said silicone polymer is a cyclic silicone polymer.

22. The cosmetic according to claim 14, wherein said silicone polymer is in the form of an aqueous solution.

23. The cosmetic according to claim 14, wherein said silicone polymer is in the form of a water dispersion.

24. The cosmetic according to claim 14, wherein said amphoteric urethane resin has at least one structural unit derived from ethylene oxide in its structure.

25. The cosmetic according to claim 14, wherein said amphoteric urethane resin has at least one polysiloxane bond in its structure.

26. The cosmetic according to claim 14, wherein said amphoteric urethane resin is in the form of an aqueous solution.

27. The cosmetic according to claim 14, which is selected from the group consisting of a hair cosmetic, a skin care cosmetic and a make-up cosmetic.



Appendix B  
(clean copy of pending claims)

14. A cosmetic comprising an amphoteric urethane resin having at least one carboxyl group and at least one tertiary amino group in a molecule, and a silicone polymer.

15. The cosmetic according to claim 14, wherein said silicone polymer is a nonionic silicone polymer.

16. The cosmetic according to claim 14, wherein said silicone polymer is a polyether-modified silicone polymer.

17. The cosmetic according to claim 14, wherein said silicone polymer is a phenyl-modified silicone polymer.

18. The cosmetic according to claim 14, wherein said silicone polymer is an amino-modified silicone polymer.

19. The cosmetic according to claim 14, wherein said silicone polymer is an alkyl-modified silicone polymer.

20. The cosmetic according to claim 14, wherein said silicone polymer is an alkoxy-modified silicone polymer.

21. The cosmetic according to claim 14, wherein said silicone polymer is a cyclic silicone polymer.

22. The cosmetic according to claim 14, wherein said silicone polymer is in the form of an aqueous solution.

23. The cosmetic according to claim 14, wherein said silicone polymer is in the form of a water dispersion.

24. The cosmetic according to claim 14, wherein said amphoteric urethane resin has at least one structural unit derived from ethylene oxide in its structure.

25. The cosmetic according to claim 14, wherein said amphoteric urethane resin has at least one polysiloxane bond in its structure.

26. The cosmetic according to claim 14, wherein said amphoteric urethane resin is in the form of an aqueous solution.

27. The cosmetic according to claim 14, which is selected from the group consisting of a hair cosmetic, a skin care cosmetic and a make-up cosmetic.

## COSMETICS

### Field of the Invention

The present invention relates to cosmetics containing an amphoteric urethane resin and a silicone  
5 polymer.

### Description of the Related Art

As a base resin for cosmetics such as hair fixatives, for example, water-soluble resins such as  
10 nonionic (non-ionic) polyvinyl pyrrolidone (PVP), cationic acrylic resin or cellulose, anionic acrylic resin or polyvinyl acetate, and amphoteric acrylic resin or polyvinyl acetate have hitherto been used. Hair fixatives comprising these water-soluble resins as the base resin  
15 have merits such as high curl holding power and excellent durability, but have problems such as poor touch, which is important for cosmetics, and drastically poor feel.

On the other hand, the present inventors found  
20 that an amphoteric urethane resin having carboxyl group(s) and tertiary amino group(s) in a molecule can provide excellent feel because of the elasticity and strong toughness of the urethane resin and filed a patent application with respect to a resin composition for  
25 cosmetics, comprising the amphoteric urethane resin as

the base resin (Japanese Patent Application No. Hei 10-27595).

However, the cosmetic comprising the amphoteric urethane resin as the base resin is superior in feel, but  
5 has such a problem that a friction coefficient of the surface is large and spread at the time of application is inferior because of poor surface smoothness.

#### Summary of the Invention

10 The present invention has been accomplished under these circumstances and an object thereof is to provide a cosmetic which is superior in both characteristics of touch and spread at the time of application.

15 To attain the object described above, the present invention is directed to a cosmetic comprising an amphoteric urethane resin having carboxyl group(s) and tertiary amino group(s) in a molecule, and a silicone polymer.

20 The present inventors have studied intensively to solve the problems such as surface smoothness, which is a weak point in case of using the amphoteric urethane resin. As a result, they have found that, when using an amphoteric urethane resin in combination with a silicone  
25 polymer, good results are obtained. That is, since the

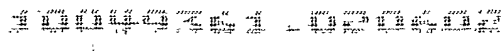


[illegible]

The cosmetics of the present invention can be obtained by using an amphoteric urethane resin having carboxyl group(s) and tertiary amino group(s) in a molecule, and a silicone polymer.

The amphoteric urethane resin having carboxyl group(s) and tertiary amino group(s) in a molecule can be prepared, for example, by reacting a polyol compound (component A), a polyisocyanate compound (component B) and a compound having active hydrogen(s) and carboxyl group(s) (component C) in the presence of excess isocyanate groups to form an isocyanate group-containing prepolymer, and reacting the isocyanate group-containing prepolymer with a compound having active hydrogen(s) and tertiary amino group(s)





polyol, polyolefin polyol and polyacrylate polyol, etc. These polyol compounds are used alone or in combination. Among these polyol compounds, polyester polyol and polyether polyol are preferably used.

5                Examples of the polyester polyol include those  
obtained by polycondensing at least one of dicarboxylic  
acids such as succinic acid, glutaric acid, adipic acid,  
sebacic acid, azelaic acid, maleic acid, fumaric acid,  
phthalic acid and terephthalic acid with at least one of  
10 polyhydric alcohols such as ethylene glycol, propylene  
glycol, 1,4-butanediol, 1,3-butanediol, 1,6-hexanediol,  
neopentyl glycol, 1,8-octanediol, 1,10-decanediol,  
diethylene glycol, spiro-glycol and trimethylolpropane,  
etc., and those obtained by the ring-opening  
15 polymerization of lactones.

Example of the polyether polyol include polyhydric alcohols used in the synthesis of the said polyester polyols, phenols such as bisphenol A, or those obtained by the ring-opening addition polymerization of primary amines or secondary amines and cyclic ether such as ethylene oxide, propylene oxide, oxetane and tetrahydrofuran. Examples thereof include polyoxyethylene polyol, polyoxypropylene polyol, polyoxytetramethylene polyol, and those obtained by the ring-opening addition polymerization of bisphenol A and

at least one of propylene oxide and ethylene oxide, etc.  
(in case of a copolymer it may be either a block copolymer or a random copolymer).

The polyisocyanate compound (component B) is  
not specifically limited, and examples thereof include  
organic diisocyanate compounds such as aliphatic  
diisocyanate compound, alicyclic diisocyanate compound  
and aromatic diisocyanate compound. These compounds  
may be used alone or in combination.

Example of the aliphatic diisocyanate compound  
includes ethylene diisocyanate, 2,2,4-trimethylhexa-  
methylenediisocyanate, 1,6-hexamethylenediisocyanate,  
etc. Examples of the alicyclic diisocyanate compound  
includes hydrogenated 4,4'-diphenylmethane  
diisocyanate, 1,4-cyclohexane diisocyanate,  
methylcyclohexylenediisocyanate, isophorone  
diisocyanate and norbornane diisocyanate, etc.  
Examples of the aromatic diisocyanate compound  
includes 4,4'-diphenylmethane diisocyanate, xylylene  
diisocyanate, toluene diisocyanate and naphthalene  
diisocyanate, etc. Among these compounds, 1,6-  
hexamethylenediisocyanate, isophorone diisocyanate,  
norbornane diisocyanate, etc. are preferable because of  
the excellent light resistance and low price.

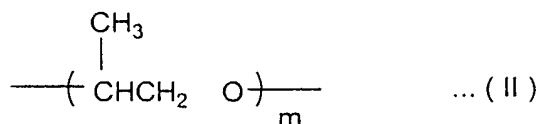
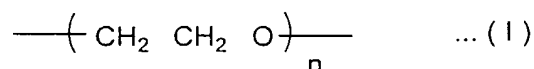
The compound (component C) having active











5           The above amphoteric urethane resin may have both EO and PO units. A proportion of the EO unit to the PO unit is preferably within a range from 10/0 to 2/8, and particularly preferably from 10/0 to 4/6, on a weight basis.

The repeating number  $n$  of the EO unit in the general formula (I) is preferably set within a range from 3 to 300, and particularly preferably from 20 to 120. When the  $n$  is less than 3, sufficient hydrophilicity can not be provided because of too small amount of the EO unit(s) to be incorporated into the amphoteric urethane resin and, therefore, sufficient hair washability are not likely to be obtained. On the other hand, when  $n$  exceeds 300, an adverse influence is likely to be exerted on the moisture resistance or the like because of too strong hydrophilicity of the amphoteric urethane resin. Furthermore, the repeating number  $m$  of the PO unit in the general formula

(II) is preferably set within a range from 3 to 300, and particularly preferably from 20 to 120. When the amphoteric urethane resin has both EO and PO units, (n + m) is preferably set within a range from 3 to 300, and particularly preferably from 20 to 120.

The amphoteric urethane resin having structural unit(s) derived from ethylene oxide (EO) can be prepared, for example, by reacting a polyol compound (component A), a polyisocyanate compound (component B), a polyethylene oxide derivatives having active hydrogen(s), and a compound having active hydrogen(s) and carboxyl group(s) (component C) in the presence of excess isocyanate groups to form an isocyanate group-containing prepolymer, and reacting the isocyanate group-containing prepolymer with a compound having active hydrogen(s) and tertiary amino group(s) (component D). Alternatively, the amphoteric urethane resin can also be prepared by replacing the sequence of the reaction between the above components C and D. As the above components A to D, the same compounds as those described above can be used.

Examples of the polyethylene oxide derivative having active hydrogen(s) to be used together with the above components A to D is not specifically limited as far as it is capable of introducing a structural unit derived

from ethylene oxide (EO) into the structure of the above amphoteric urethane resin, and examples thereof include polyoxyethylene glycol (PEG), polyoxyethylene polyoxypropylene glycol (EOPO block copolymer), etc.

- 5 Among these, polyoxyethylene glycol is preferably used. The above polyoxyethylene oxide derivative may be any of a type wherein an OH group is introduced at both ends, a type wherein an NH<sub>2</sub> group is introduced at both ends, a type wherein an OH group is introduced at one end, and a type wherein an NH<sub>2</sub> group is introduced at one end. When using the type wherein an OH group is introduced at both ends or a type wherein an NH<sub>2</sub> group is introduced at both ends, an amphoteric urethane resin having the EO unit(s) in a principal chain is obtained.
- 10
- 15 When using the type wherein an OH group is introduced at one end or a type wherein an NH<sub>2</sub> group is introduced at one end, an amphoteric urethane resin having EO unit(s) at its side chain(s) or end(s) is obtained.

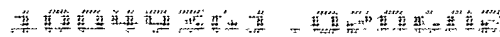
- 20 The molecular weight of the specific polyethylene oxide derivative is preferably within a range from 200 to 20000, and particularly preferably from 1000 to 10000

To further improve the feel, those having polysiloxane bond(s) in the structure are preferably used as the amphoteric urethane resin.

- 25 The repeating number n of a siloxane bond (Si-O)

is preferably within a range from 5 to 300, and particularly preferably from 20 to 150. When n is less than 5, it becomes difficult to obtain the sufficient effect on the touch, coming properties, etc. obtained intrinsically by introducing the polysiloxane bond(s) because of too small amount of the polysiloxane bond(s) in the resulting amphoteric urethane resin. On the other hand, when n exceeds 300, the compatibility with other raw materials become inferior because of high hydrophobicity, thereby making it difficult to react them. Furthermore, the adhesion to hair is likely to be inhibited because of too high hydrophobicity of the resulting polymer.

The amphoteric urethane resin having the said polysiloxane bond(s) can be prepared, for example, by reacting a polyol compound (component A), a polyisocyanate compound (component B), a polysiloxane compound having active hydrogen(s) and a compound having active hydrogen(s), and carboxyl group(s) (component C) in the presence of excess isocyanate groups to form an isocyanate group-containing prepolymer, and reacting the isocyanate group-containing prepolymer with a compound having active hydrogen(s) and tertiary amino group(s) (component D). Alternatively, the amphoteric urethane resin can also be



prepared by replacing the sequence of the reaction between the above components C and D. As the above components A to D, the same compounds as those described above can be used.

5           The polysiloxane compound having active hydrogen(s) to be used together with the above components A to D is not specifically limited as far as it is capable of introducing polysiloxane bond(s) into the structure of the amphoteric urethane resin, and examples thereof include polydialkylsiloxanediol, polydialkylsiloxanemonool, polydialkylsiloxanediamine, polydialkylsiloxanemonoamine, etc. These compounds may be used alone or in combination. The alkyl group(s) to be combined with Si of the respective siloxane bonds of the said polydialkylsiloxanediol preferably has 1 to 10 carbon atoms, and particularly preferably 1 to 5 carbon atoms. The above polysiloxane compound may contain various siloxane bonds wherein the number of carbon atoms of the alkyl group(s) to be combined with Si of the siloxane bonds varies. Specific examples of the polydialkylsiloxanediol include polydimethylsiloxanediol, polymethylethylsiloxanediol, etc. Examples of the polydialkylsiloxanemonool include polydimethylsiloxanemonool, polymethylethylsiloxanemonool, etc. Examples of the polydialkylsiloxanediamine include polydimethylsiloxane-



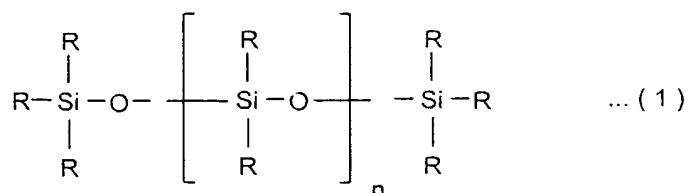


where the amphoteric urethane resin is dispersed in water.

It is also possible to add a crosslinking agent such as silane coupling agent to the water dispersion of the above amphoteric resin, thereby to provide the crosslinkability. To provide the storage stability, various additives may be added and examples thereof include protective colloidal agents, bactericides, mildewproofing agents, etc.

The silicone polymer used together with the said amphoteric urethane resin is not specifically limited as far as it has polysiloxane bond(s) in a molecule and can be used in cosmetics, and examples thereof include silicone resin, silicone oil, silicone emulsion, silicone rubber, etc. Examples of the silicone rubber include nonionic, polyether-modified, phenyl-modified, amino-modified, alkyl-modified, alkoxy-modified, cyclic silicone polymers, etc. These silicone polymers can be used alone or in combination.

Examples of the nonionic silicone polymer includes those represented by the following general formula (1):



[wherein R represents a hydrogen atom, hydrocarbon group(s) having 1 to 12 carbon atoms, or  $-\text{OSi}(\text{CH}_3)_3$  and may be the same or different; and n represents a numeral of 3 or more].

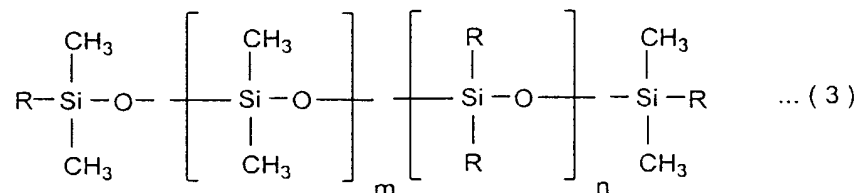
In the general formula (1), the hydrocarbon group(s) having 1 to 12 carbon atoms is/are straight-chain or branched-chain saturated hydrocarbon group(s) and is/are preferably methyl group(s), while n is preferably from 50 to 3000.

Examples of the nonionic silicone polymer represented by the general formula (1) include methyl polysiloxane whose R is entirely methyl group(s).

Examples of the polyether-modified silicone polymer includes those represented by the following general formula (2). Each repeating unit in the silicone polymer of the present invention may be in any form of polymerization such as random polymerization and block polymerization.



includes those represented by the following general formula (3):

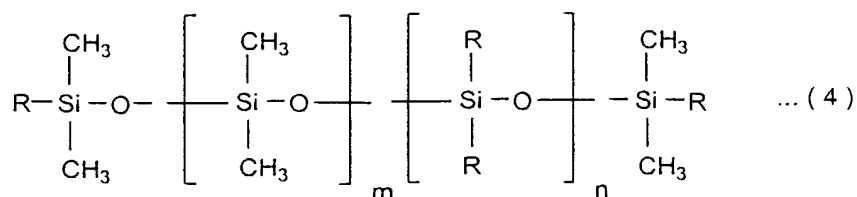


5 [wherein R represents hydrocarbon group(s) having 1 to 12 carbon atoms,  $-\text{OSi}(\text{CH}_3)_3$  or phenyl group(s) and may be the same or different, provided that at least one of R(s) is phenyl group(s); m represents 0 or a numeral of 1 or more; and n represents a numeral of 1 or more].

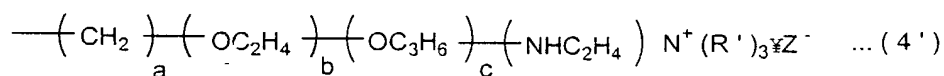
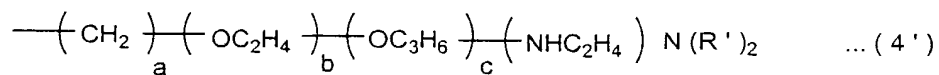
10 In the general formula (3), the hydrocarbon group(s) having 1 to 12 carbon atoms represented by R include(s) straight-chain or branched-chain saturated hydrocarbon group(s). m is preferably from 0 to 500, and n is preferably from 1 to 2000.

15 As the phenyl-modified silicone polymer represented by the general formula (3) methylphenyl polysiloxane is preferably used.

Examples of the amino-modified silicone polymer includes those represented by the following general  
20 formula (4):



[wherein R represents a hydrogen atom, hydrocarbon group(s) having 1 to 12 carbon atoms, hydroxyl group(s),  
 5 methoxy group(s) or group(s) represented by the following general formula (4') or (4'') and may be the same or different, provided that at least one of R(s) is group(s) represented by the following general formula (4') or (4''); m represents 0 or a numeral of 1 or more;  
 10 and n represents a numeral of 1 or more

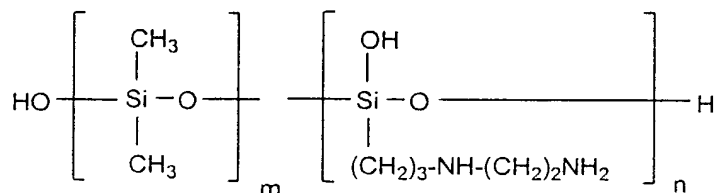


(wherein R' represents a hydrogen atom or hydrocarbon group(s) having 1 to 6 carbon atoms and may be the same or different; Z represents a halogen ion or an  
 15 organic anion; a represents a numeral of 1 to 6; b represents a numeral of 0 to 6; and c represents a numeral of 0 to 6)].

In the general formula (4), the hydrocarbon  
 20 group(s) having 1 to 12 carbon atoms represented by R

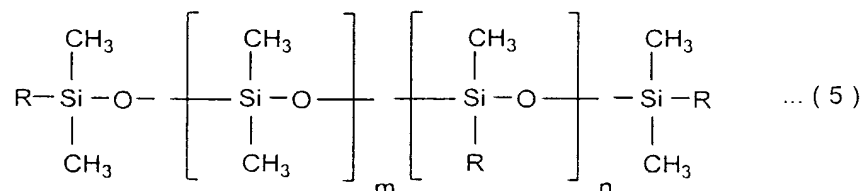
include(s) straight-chain or branched-chain saturated hydrocarbon group(s). m is preferably from 3 to 500, and n is preferably from 1 to 500. In the general formulas (4') and (4''), the hydrocarbon group(s) having 1 to 6 carbon atoms represented by R' include(s) straight-chain or branched-chain saturated hydrocarbon group(s).

As the amino-modified silicone polymer represented by the said general formula (4), aminodimethicone represented by the following structural formula is preferably used.



[wherein m and n are as defined in the general formula (4)]

Examples of the alkyl-modified silicone polymer include, those represented by the following general formula (5):

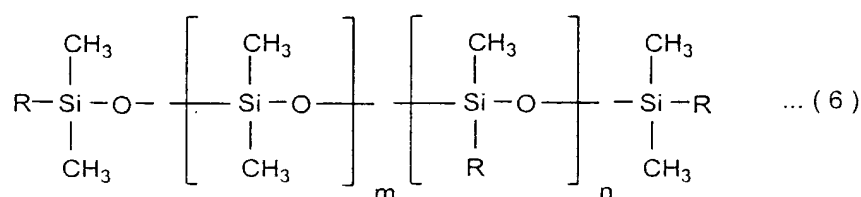


[wherein R represents hydrocarbon group(s) having 1 to

50 carbon atoms and may be the same or different, provided that at least one of R(s) is hydrocarbon group(s) having 12 to 50 carbon atoms; m represents 0 or a numeral of 1 or more; and n represents a numeral of 1 or more].

In the general formula (5), the hydrocarbon group(s) having 1 to 50 carbon atoms represented by R include(s) straight-chain or branched-chain saturated hydrocarbon group(s), and hydrocarbon group(s) having 12 to 50 carbon atoms is/are preferably used, and hydrocarbon group(s) having 15 to 40 carbon atoms is/are more preferably used. m is preferably from 10 to 500, and n is preferably from 10 to 500.

Examples of the alkoxy-modified silicone polymer include those represented by the following general formula (6):



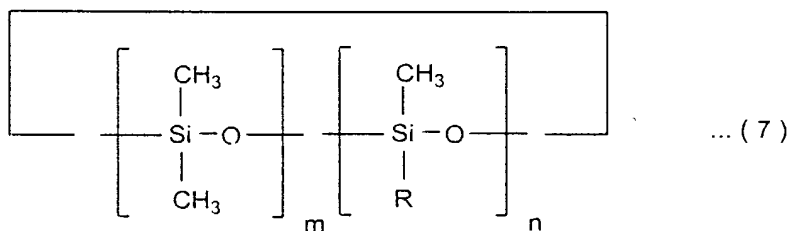
[wherein R represents hydrocarbon group(s) having 1 to 12 carbon atoms or alkoxy group(s) having 1 to 50 carbon atoms and may be the same or different, provided that at least one of R(s) is alkoxy group(s) having 1 to 50



carbon atoms; m represents 0 or a numeral of 1 or more; and n represents a numeral of 1 or more].

In the general formula (6), the hydrocarbon group(s) having 1 to 12 carbon atoms represented by R include(s) straight-chain or branched-chain saturated hydrocarbon group(s). The alkoxy group(s) having 1 to 50 carbon atoms represented by R include(s) straight-chain or branched-chain saturated alkoxy group(s) and alkoxy group(s) having 1 to 20 carbon atoms is/are preferably used. m is preferably from 3 to 500, and n is preferably from 1 to 100.

Examples of the cyclic silicone polymer includes those represented by the following general formula (7):



[wherein R represents hydrocarbon group(s) having 1 to 12 carbon atoms and may be the same or different in each repeating unit; m represents 0 or a numeral of 1 or more; n represents a numeral of 1 or more; and m + n is from 4 to 10].

In the general formula (7), the hydrocarbon group(s) having 1 to 12 carbon atoms represented by R



urethane resin thus obtained described above, a silicone polymer, various surfactants such as polyoxyethylene alkyl ether, and coconut oil fatty acid diethanolamide, ethanol, deionized water, etc. are blended in the predetermined proportion, and mixed until they are made homogenous to obtain a component X. Then, a component Y made of a propellant (LPG) is added to prepare a desired foam hair fixative.

#### Preparation of hair cosmetics (aerosol spray hair fixative)

In the aqueous solution of the above amphoteric urethane resin, a silicone polymer, deionized water, sodium dioctyl sulfosuccinate, ethanol, etc. are blended in the predetermined proportion, and mixed until they are made homogenous to obtain a component X. Then, a component Y made of a propellant (LPG) is added to prepare a desired aerosol spray hair fixative.

#### Preparation of hair cosmetics (gel-like hair fixatives)

First, a viscosity modifier, triethanolamine, ethanol, deionized water, etc. are blended in the predetermined proportion, and then mixed until a viscous gel is formed to obtain a component X. Then, a silicone polymer, ethanol, deionized water, etc. are blended in the aqueous solution of the amphoteric urethane resin in the predetermined proportion to obtain a component Y. The resulting component Y is added to the above component,

and mixed until they are made homogenous to prepare a desired gel-like hair fixative.

Preparation of hair cosmetics (pump spray hair fixatives)

In the aqueous solution of the amphoteric urethane resin, a silicone polymer, sodium dioctyl sulfosuccinate, ethanol, deionized water, etc. are blended in the predetermined proportion, and mixed until they are made homogenous to prepare a desired pump spray hair fixative.

Furthermore, skin care cosmetics such as shaving cream, skin care lotion and sunscreen lotion; and make-up cosmetics such as nail polish, mascara and foundation can be prepared according to the general preparation methods of these cosmetics.

Examples

The following Examples and Comparative Examples further illustrate the present invention in detail.

The following materials were prepared before describing the Examples and Comparative Examples.

Amphoteric urethane resin (a)

In a glass four-necked flask equipped with a stirrer, a thermometer, a nitrogen introducing tube and a reflux condenser, 100 g of isophorone diisocyanate (IPDI), 60 g of polypropylene glycol (PPG having a molecular weight of 1000), 5 g of cyclohexyl dimethanol

(CHDM), and 38 g of dimethylolbutanoic acid (DMBA) were charged, and then 60 g of ethyl acetate as a solvent was added and the mixture was heated to 80 degree C in an oil bath and allowed to react for four hours. Then, 2 g of N-methyldiethanolamine and 30 g of ethyl acetate were added furthermore, and the mixture was allowed to react for additional three hours. To the resulting mixture, 30 g of polypropylene glycol having an amino group at one end (Jeffamine M1000, manufactured by HUNTSMAN CORPORATION) and 50 g of ethyl acetate were added furthermore, and the mixture was allowed to react for additional one hour to obtain a solution of a polyurethane prepolymer having residual NCO groups. The polyurethane prepolymer having residual NCO groups was dispersed in 750 g of water containing 16 g of potassium hydroxide and then polymerized by the chain-extending reaction at 50 degree C for three hours. Ethyl acetate was recovered from the resulting water dispersion under reduced pressure to obtain an amphoteric urethane resin which did not substantially contain the solvent.

#### Amphoteric urethane resin (b)

In a glass four-necked flask equipped with a stirrer, a thermometer, a nitrogen introducing tube and a reflux condenser, 100 g of isophorone diisocyanate



substantially contain the solvent and had ethylene oxide chain(s) in the structure.

Amphoteric urethane resin (c)

In a glass four-necked flask equipped with a stirrer, a thermometer, a nitrogen introducing tube and a reflux condenser, 100 g of isophorone diisocyanate (IPDI), and 3 g of polydimethylsiloxanediol having two OH groups at one end (molecular weight: 1000) were charged, and then the mixture was heated to 80 degree C in an oil bath and allowed to react for two hours. Then, 55 g of polypropylene glycol (PPG having a molecular weight of 1000), 10 g of hydrogenated bisphenol A, and 36 g of dimethylolbutanoic acid (DMBA) were added, and then 60 g of ethyl acetate as a solvent was added, and the mixture was heated to 80 degree C in an oil bath and allowed to react for four hours. Then, 2 g of N-methyldiethanolamine and 30 g of ethyl acetate were added furthermore, and the mixture was allowed to react for additional three hours. To the resulting mixture, 30 g of polypropylene glycol having an amino group at one end (Jeffamine M1000, manufactured by HUNTSMAN CORPORATION) and 50 g of ethyl acetate were added furthermore, and the mixture was allowed to react for additional one hour to obtain a solution of a polyurethane prepolymer having residual NCO groups. The

polyurethane prepolymer having residual NCO groups was dispersed in 750 g of water containing 15 g of potassium hydroxide and then polymerized by the chain-extending reaction at 50 degree C for three hours. Ethyl acetate was recovered from the resulting water dispersion under reduced pressure to obtain an aqueous substance of an amphoteric urethane resin which did not substantially contain the solvent and had dimethylsiloxane chain(s) in the structure.

10 Silicone polymer (1) (nonionic)

Methylpolysiloxane (SH200C-2 manufactured by DOW CORNING TORAY SILICONE CO., LTD.)

Silicone polymer (2) (polyether-modified)

SH3771C manufactured by DOW CORNING  
15 TORAY SILICONE CO., LTD.

Silicone polymer (3) (phenyl-modified)

Methylphenylpolysiloxane (SH556 manufactured by DOW CORNING TORAY SILICONE CO., LTD.)

Silicone polymer (4) (amino-modified)

20 Amodimethicone (SM8702C manufactured by DOW CORNING TORAY SILICONE CO., LTD.)

Silicone polymer (5) (alkyl-modified)

KF-412 manufactured by SHIN-ETSU CHEMICAL CO., LTD.



$$\frac{1}{\sqrt{\pi}} \left( \frac{1}{\sqrt{\pi}} \right)^n = \frac{1}{\sqrt{\pi}^n}$$

Silicone polymer (7) (cyclic)

Polyoxyethylene stearyl ether

10 Coconut oil fatty acid diethanolamide

Sodium dioctyl sulfosuccinate

Viscosity modifier

20 Hair cosmetics (foam hair fixatives)

Examples 1a to 21a. Comparative Examples 1a to 3a

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### Touch

0.8 g of the foam hair fixative was applied on a strand of black virgin hairs (having a length of 25 cm and a weight of 5.0 g). Then, the strand of hairs after drying at room temperature was subjected to an organoleptic test using ten panelists and the touch for hair cosmetic was evaluated. Evaluation criteria are set as follows:

: The number of persons, who felt that the strand of hairs after drying is very smooth to the touch, is 9 or more.

: The number of persons, who felt that the strand of hairs after drying is very smooth to the touch, is within a range from 6 to 8.

: The number of persons, who felt that the strand of hairs after drying is very smooth to the touch, is within a range from 2 to 5.

×: The number of persons, who felt that the strand of hairs after drying is very smooth to the touch, is 1 or less.

### 20 Hair washability

0.6 g of the foam hair fixative was applied to black virgin hairs (having a length of 15 cm and a weight of 3 g), followed by drying to make a strand of hairs. After the strand of hairs was slightly loosen using hot water at



Table 1  
(Foam hair Fixative)  
(Part)

	Examples						
	1a	2a	3a	4a	5a	6a	7a
Component X	Amphoteric urethane resin (Types)	3.0 (a)	3.0 (a)	3.0 (a)	3.0 (a)	3.0 (a)	3.0 (a)
	Silicone polymer (Types)	0.5 (1)	0.5 (2)	0.5 (3)	0.5 (4)	0.5 (5)	0.5 (6)
	Deionized water	77.2	77.2	77.2	77.2	77.2	77.2
	Polyoxyethylene stearyl ether	0.5	0.5	0.5	0.5	0.5	0.5
Component Y	Ethanol	10.0	10.0	10.0	10.0	10.0	10.0
	Coconut oil fatty acid diethanolamide	0.8	0.8	0.8	0.8	0.8	0.8
	Propellant (LPG)	8.0	8.0	8.0	8.0	8.0	8.0
Feel							
Spread							
Touch							
Hair washability							

Table 2

(Foam Hair Fixative) (Part)

	Examples									
	8a	9a	10a	11a	12a	13a	14a			
Component X	Amphoteric urethane resin (Types)	3.0 (b)	3.0 (b)	3.0 (b)	3.0 (b)	3.0 (b)	3.0 (b)	3.0 (b)	3.0 (b)	3.0 (b)
	Silicone polymer (Types)	0.5 (1)	0.5 (2)	0.5 (3)	0.5 (4)	0.5 (5)	0.5 (6)	0.5 (7)	0.5 (7)	0.5 (7)
	Deionized water	77.2	77.2	77.2	77.2	77.2	77.2	77.2	77.2	77.2
	Polyoxyethylene stearyl ether	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5
Component Y	Ethanol	10.0	10.0	10.0	10.0	10.0	10.0	10.0	10.0	10.0
	Coconut oil fatty acid diethanolamide	0.8	0.8	0.8	0.8	0.8	0.8	0.8	0.8	0.8
Component Y	Propellant (LPG)	8.0	8.0	8.0	8.0	8.0	8.0	8.0	8.0	8.0
Feel										
Spread										
Touch										
Hair washability										

Table 3

(Foam Hair Fixative) (Part)

	Examples									
	15a	16a	17a	18a	19a	20a	21a			
Component X	Amphoteric urethane resin (Types)	3.0 (c)	3.0 (c)	3.0 (c)	3.0 (c)	3.0 (c)	3.0 (c)			
	Silicone polymer (Types)	0.5 (1)	0.5 (2)	0.5 (3)	0.5 (4)	0.5 (5)	0.5 (6)			
	Deionized water	77.2	77.2	77.2	77.2	77.2	77.2			
	Polyoxyethylene stearyl ether	0.5	0.5	0.5	0.5	0.5	0.5			
	Ethanol	10.0	10.0	10.0	10.0	10.0	10.0			
Component Y	Coconut oil fatty acid diethanolamide	0.8	0.8	0.8	0.8	0.8	0.8			
	Propellant (LPG)	8.0	8.0	8.0	8.0	8.0	8.0			
Feel										
Spread										
Touch										
Hair washability										



Table 4  
(Foam Hair Fixative)  
(Product)

	Comparative Examples		
	1a	2a	3a
Component X	Amphoteric urethane resin (Types)	3.0 (b)	3.0 (c)
	Silicone polymer (Types)	—	—
	Deionized water	77.7	77.7
	Polyoxyethylene stearyl ether	0.5	0.5
	Ethanol	10.0	10.0
Component Y	Coconut oil fatty acid diethanolamide	0.8	0.8
	Propellant (LPG)	8.0	8.0
Feel			
Spread			
Touch			
Hair washability			

As is apparent from the results shown in Tables  
 1 to 4, the foam hair fixatives of the Examples have very  
 good feel and good spread at the time of application and  
 are superior in touch and hair washability because the  
 5 amphoteric urethane resin and silicone polymer are used  
 in combination. It is also apparent that the foam hair  
 fixatives of the Examples using the amphoteric urethane  
 resin (b) having ethylene oxide chain(s) in its structure  
 are markedly superior in hair washability. It is also  
 10 apparent that the foam hair fixatives of the Examples  
 using the amphoteric urethane resin (c) having  
 polysiloxane bonds in its structure are markedly superior  
 in touch.

To the contrary, the foam hair fixatives of the  
 15 Comparative Examples have good feel because the  
 amphoteric urethane resin is used, but are inferior in  
 spread at the time of application.

#### Hair cosmetics (aerosol spray hair fixatives)

#### Examples 1b to 21b, Comparative Examples 1b to 3b

20 The respective materials of a component X shown  
 in Tables 5 to 8 described hereinafter were blended in  
 the proportion shown in the same tables and mixed until  
 they are made homogenous to obtain the component X.  
 Then, a component Y was added in the resulting



Table 5  
(Aerosol spray hair fixative)  
(Parts)

	Examples						
	1b	2b	3b	4b	5b	6b	7b
Component X	Amphoteric urethane resin (Types)	3.0 (a)	3.0 (a)	3.0 (a)	3.0 (a)	3.0 (a)	3.0 (a)
	Silicone polymer (Types)	0.5 (1)	0.5 (2)	0.5 (3)	0.5 (5)	0.5 (6)	0.5 (7)
	Deionized water	7.0	7.0	7.0	7.0	7.0	7.0
	Sodium dioctyl sulfosuccinate	0.3	0.3	0.3	0.3	0.3	0.3
	Ethanol	49.2	49.2	49.2	49.2	49.2	49.2
Component Y	Propellant (LPG)	40.0	40.0	40.0	40.0	40.0	40.0
Feel							
Spread							
Touch							
Hair washability							

Table 6  
(Aerosol spray hair fixative) (Parts)

	Examples									
	8b	9b	10b	11b	12b	13b	14b			
Component X	Amphoteric urethane resin (Types)	3.0 (b)	3.0 (b)	3.0 (b)	3.0 (b)	3.0 (b)	3.0 (b)	3.0 (b)	3.0 (b)	3.0 (b)
	Silicone polymer (Types)	0.5 (1)	0.5 (2)	0.5 (3)	0.5 (4)	0.5 (5)	0.5 (6)	0.5 (6)	0.5 (7)	0.5 (7)
	Deionized water	7.0	7.0	7.0	7.0	7.0	7.0	7.0	7.0	7.0
	Sodium dioctyl sulfosuccinate	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3
	Ethanol	49.2	49.2	49.2	49.2	49.2	49.2	49.2	49.2	49.2
Component Y	Propellant (LPG)	40.0	40.0	40.0	40.0	40.0	40.0	40.0	40.0	40.0
Feel										
Spread										
Touch										
Hair washability										

Table 7  
(Aerosol spray hair fixative)  
(Parts)

		Examples						
		15b	16b	17b	18b	19b	20b	21b
Component X	Amphoteric urethane resin (Types)	3.0 (c)	3.0 (c)	3.0 (c)	3.0 (c)	3.0 (c)	3.0 (c)	3.0 (c)
	Silicone polymer (Types)	0.5 (1)	0.5 (2)	0.5 (3)	0.5 (4)	0.5 (5)	0.5 (6)	0.5 (7)
	Deionized water	7.0	7.0	7.0	7.0	7.0	7.0	7.0
	Sodium dioctyl sulfosuccinate	0.3	0.3	0.3	0.3	0.3	0.3	0.3
	Ethanol	49.2	49.2	49.2	49.2	49.2	49.2	49.2
Component Y	Propellant (LPG)	40.0	40.0	40.0	40.0	40.0	40.0	40.0
Feel								
Spread								
Touch								
Hair washability								

Table 8  
(Aerosol spray hair fixative)  
(Parts)

	Comparative Examples		
	1b	2b	3b
Component X	Amphoteric urethane resin (Types)	3.0 (b)	3.0 (c)
	Silicone polymer (Types)	-	-
	Deionized water	7.0	7.0
	Sodium dioctyl sulfosuccinate	0.3	0.3
	Ethanol	49.7	49.7
Component Y		40.0	40.0
Propellant (LPG)		40.0	40.0
Feel			
Spread			
Touch			
Hair washability			





table was added in the resulting component X and mixed until they are made homogenous to prepare a jelly hair fixative, respectively.

Using the jerry hair fixatives of the Examples and  
 5 Comparative Examples thus obtained, the respective characteristics were evaluated according to the criteria for hair cosmetics. The results are summarized in Tables 9 to 12 described hereinafter.

Table 9

(Parts)

(Gel-like hair fixative)

	Examples						
	1c	2c	3c	4c	5c	6c	7c
Component X	Thickener	1.5	1.5	1.5	1.5	1.5	1.5
	Triethanolamine	1.1	1.1	1.1	1.1	1.1	1.1
	Ethanol	5.0	5.0	5.0	5.0	5.0	5.0
	Deionized water	50.0	50.0	50.0	50.0	50.0	50.0
Component Y	Amphoteric urethane resin (Types)	3.0 (a)	3.0 (a)	3.0 (a)	3.0 (a)	3.0 (a)	3.0 (a)
	Silicone polymer (Types)	0.5 (1)	0.5 (2)	0.5 (3)	0.5 (4)	0.5 (5)	0.5 (6)
	Ethanol	5.0	5.0	5.0	5.0	5.0	5.0
	Deionized water	33.9	33.9	33.9	33.9	33.9	33.9
Feel							
Spread							
Touch							
Hair washability							

Table 10

(Gel-like hair fixative) (Parts)

		Examples						
		8c	9c	10c	11c	12c	13c	14c
Component X	Thickener	1.5	1.5	1.5	1.5	1.5	1.5	1.5
	Triethanolamine	1.1	1.1	1.1	1.1	1.1	1.1	1.1
	Ethanol	5.0	5.0	5.0	5.0	5.0	5.0	5.0
	Deionized water	50.0	50.0	50.0	50.0	50.0	50.0	50.0
Component Y	Amphoteric urethane resin (Types)	3.0 (b)	3.0 (b)	3.0 (b)	3.0 (b)	3.0 (b)	3.0 (b)	3.0 (b)
	Silicone polymer (Types)	0.5 (1)	0.5 (2)	0.5 (3)	0.5 (4)	0.5 (5)	0.5 (6)	0.5 (7)
	Ethanol	5.0	5.0	5.0	5.0	5.0	5.0	5.0
	Deionized water	33.9	33.9	33.9	33.9	33.9	33.9	33.9
Feel								
Spread								
Touch								
Hair washability								

Table 11

(Gel-like hair fixative) (Parts)

	Examples									
	15c	16c	17c	18c	19c	20c	21c			
Component X	Thickener	1.5	1.5	1.5	1.5	1.5	1.5			
	Triethanolamine	1.1	1.1	1.1	1.1	1.1	1.1			
	Ethanol	5.0	5.0	5.0	5.0	5.0	5.0			
	Deionized water	50.0	50.0	50.0	50.0	50.0	50.0			
Component Y	Amphoteric urethane resin (Types)	3.0 (c)	3.0 (c)	3.0 (c)	3.0 (c)	3.0 (c)	3.0 (c)			
	Silicone polymer (Types)	0.5 (1)	0.5 (2)	0.5 (3)	0.5 (4)	0.5 (5)	0.5 (6)			
	Ethanol	5.0	5.0	5.0	5.0	5.0	5.0			
	Deionized water	33.9	33.9	33.9	33.9	33.9	33.9			
Feel										
Spread										
Touch										
Hair washability										

		Comparative Examples		
		1c	2c	3c
Component X	Thickener	1.5	1.5	1.5
	Triethanolamine	1.1	1.1	1.1
	Ethanol	5.0	5.0	5.0
	Deionized water	50.0	50.0	50.0
	Amphoteric urethane resin (Types)	3.0	3.0	3.0
Component Y	Silicone polymer (Types)	-	-	-
	Ethanol	5.0	5.0	5.0
	Deionized water	34.4	34.4	34.4
Feel				
Spread				
Touch				
Hair washability				

Table 12 (Gel-like hair fixative) (Parts)

As is apparent from the results shown in Tables 9 to 12, the jelly hair fixatives of the Examples have very good feel and good spread at the time of application and are superior in touch and hair washability because the amphoteric urethane resin and silicone polymer are used in combination. It is also apparent that the gel-like hair fixatives of the Examples using the amphoteric urethane resin (b) having ethylene oxide chain(s) in its structure are markedly superior in hair washability. It is also apparent that the gel-like hair fixatives of the Examples using the amphoteric urethane resin (c) having polysiloxane bond(s) in its structure are markedly superior in touch.

To the contrary, the gel-like hair fixatives of the Comparative Examples have good feel because the amphoteric urethane resin is used, but are inferior in spread at the time of application.

Hair cosmetics (pump spray hair fixatives)

Examples 1d to 21d, Comparative Examples 1d to 3d

20           The respective materials shown in Tables 13 to 16 described hereinafter were blended in the proportion shown in the same table and mixed until they are made homogenous to prepare a pump spray hair fixative, respectively.



Table 13  
(Pump spray hair fixative) (Parts)

	Examples						
	1d	2d	3d	4d	5d	6d	7d
Amphoteric urethane resin (Types)	3.0 (a)	3.0 (a)	3.0 (a)	3.0 (a)	3.0 (a)	3.0 (a)	3.0 (a)
Silicone polymer (Types)	0.5 (1)	0.5 (2)	0.5 (3)	0.5 (4)	0.5 (5)	0.5 (6)	0.5 (7)
Sodium dioctyl sulfosuccinate	0.3	0.3	0.3	0.3	0.3	0.3	0.3
Ethanol	10.0	10.0	10.0	10.0	10.0	10.0	10.0
Deionized water	86.2	86.2	86.2	86.2	86.2	86.2	86.2
Feel							
Spread							
Touch							
Hair washability							



Table 14

(Parts)

(Pump spray hair fixative)

	Examples							
	8d	9d	10d	11d	12d	13d	14d	
Amphoteric urethane resin (Types)	3.0 (b)	3.0 (b)	3.0 (b)	3.0 (b)	3.0 (b)	3.0 (b)	3.0 (b)	
Silicone polymer (Types)	0.5 (1)	0.5 (2)	0.5 (3)	0.5 (4)	0.5 (5)	0.5 (6)	0.5 (7)	
Sodium dioctyl sulfosuccinate	0.3	0.3	0.3	0.3	0.3	0.3	0.3	
Ethanol	10.0	10.0	10.0	10.0	10.0	10.0	10.0	
Deionized water	86.2	86.2	86.2	86.2	86.2	86.2	86.2	
Feel								
Spread								
Touch								
Hair washability								

Table 15

(Pump spray hair fixative) (Parts)

	Examples									
	15d	16d	17d	18d	19d	20d	21d			
Ampholeric urethane resin (Types)	3.0 (c)	3.0 (c)	3.0 (c)	3.0 (c)	3.0 (c)	3.0 (c)	3.0 (c)			
Silicone polymer (Types)	0.5 (1)	0.5 (2)	0.5 (3)	0.5 (4)	0.5 (5)	0.5 (6)	0.5 (7)			
Sodium dioctyl sulfosuccinate	0.3	0.3	0.3	0.3	0.3	0.3	0.3			
Ethanol	10.0	10.0	10.0	10.0	10.0	10.0	10.0			
Deionized water	86.2	86.2	86.2	86.2	86.2	86.2	86.2			
Feel										
Spread										
Touch										
Hair washability										

Table 16  
(Pump spray hair fixative)  
(Parts)

	Comparative Examples		
	1d	2d	3d
Ampholeric urethane resin (Types)	3.0 (a)	3.0 (b)	3.0 (c)
Silicone polymer (Types)	— —	— —	— —
Sodium dioctyl sulfosuccinate	0.3	0.3	0.3
Ethanol	10.0	10.0	10.0
Deionized water	86.7	86.7	86.7
Feel			
Spread			
Touch			
Hair washability			



degree C to obtain the component Y. Then, the component X and component Y were mixed, followed by stirring at 80 degree C for 30 minutes. A viscosity modifier was added, and mixed until they were made  
 5 homogenous, then cooled down to 40 degree C to prepare a skin care lotion, respectively.

Using the skin care lotions of the Examples and Comparative Example thus obtained, the respective characteristics were evaluated according to the following  
 10 criteria. The results are summarized in Tables 17 and 18 described hereinafter.

#### Feel

The feel for skin care cosmetic was evaluated at a practical use test using ten panelists. Evaluation criteria  
 15 are set as follows:

: The number of persons, who did not feel tight and also felt soft, is 9 or more.

: The number of persons, who did not feel tight and also felt soft, is within a range from 6 to 8

20 : The number of persons, who did not feel tight and also felt soft, is within a range from 2 to 5.

×: The number of persons, who did not feel tight and also felt soft, is 1 or less

Spread

The spread for skin care cosmetic was evaluated by a practical use test using ten panelists. Evaluation criteria are set as follows:

5                   : The number of persons, who felt that the spread at the time of application is very good, is 9 or more.

10 : The number of persons, who felt that the spread at the time of application is very good, is within a range from 6 to 8.

: The number of persons, who felt that the spread at the time of application is very good, is within a range from 2 to 5.

15       ×: The number of persons, who felt that the spread at the time of application is very good, is 1 or less.

Table 17

(Skin care lotion) (Parts)

	Examples			
	1e	2e	3e	4e
Component X	Octyl methoxycinnamate	7.5	7.5	7.5
	Ether polyoxystearate	1.0	1.0	1.0
	Emulsion type glyceryl stearate	1.0	1.0	1.0
	Stearic acid	1.5	1.5	1.5
	Mixture of titanium dioxide and C <sub>12-15</sub> alkyl benzoate	1.7	1.7	1.7
Component Y	Polyoxyethylene-added dimethylcone	0.5	0.5	0.5
	Amphoteric urethane resin (a)	1.0	1.0	1.0
	Silicone polymer (Types)	0.3 (1)	0.3 (2)	0.3 (3) (4)
	Deionized water	61.5	61.5	61.5
	Triethanolamine (99%)	4.0	4.0	4.0
Feel	Antiseptic	q.s.	q.s.	q.s.
	Thickener (2%)	20.0	20.0	20.0
Spread				

Table 18

(Parts)

(Skin care lotion)

	Examples	Comparative Example		
		5e	6e	7e
Component X	Octyl methoxycinnamate	7.5	7.5	7.5
	Ether polyoxystearate	1.0	1.0	1.0
	Emulsion type glyceryl stearate	1.0	1.0	1.0
	Stearic acid	1.5	1.5	1.5
	Mixture of titanium dioxide and C <sub>12-15</sub> alkyl benzoate	1.7	1.7	1.7
Component Y	Polyoxyethylene-added dimethicone	0.5	0.5	0.5
	Amphoteric urethane resin (a)	1.0	1.0	1.0
	Silicone polymer (Types)	0.3 (5)	0.3 (6)	0.3 (7)
	Deionized water	61.5	61.5	61.5
	Triethanolamine (99%)	4.0	4.0	4.0
Feel	Antiseptic	q s.	q s.	q s.
	Thickener (2%)	20.0	20.0	20.0
	Spread			







Table 19  
(Shaving cream) (Parts)

	Examples			
	1f	2f	3f	4f
Component X	8.0	8.0	8.0	8.0
	2.0	2.0	2.0	2.0
	2.0	2.0	2.0	2.0
	0.5	0.5	0.5	0.5
	0.5	0.5	0.5	0.5
Component Y	0.3	0.3	0.3	0.3
	(1)	(2)	(3)	(4)
	72.5	72.5	72.5	72.5
	10.0	10.0	10.0	10.0
	4.2	4.2	4.2	4.2
Feel				
Spread				

Table 20

(Shaving cream)		(Parts)				Comparative Example
		Examples				
		5f	6f	7f	1f	
Component X	Stearic acid	8.0	8.0	8.0	8.0	
	Mineral oil	2.0	2.0	2.0	2.0	
	Isopropyl myristate	2.0	2.0	2.0	2.0	
	Glyceryl stearate	0.5	0.5	0.5	0.5	
Component Y	Amphoteric urethane resin (a)	0.5	0.5	0.5	0.5	
	Silicone polymer (Types)	0.3 (5)	0.3 (6)	0.3 (7)	—	
	Deionized water	72.5	72.5	72.5	72.8	
	Thickeners (2%)	10.0	10.0	10.0	10.0	
	Triethanolamine (99%)	4.2	4.2	4.2	4.2	
Feel						
Spread						

As is apparent from the results shown in Tables 19 and 20, the shaving creams of the Examples have very good feel and good spread at the time of application because the amphoteric urethane resin and silicone polymer are used in combination.

To the contrary, the shaving cream of the Comparative Example has good feel because the amphoteric urethane resin is used, but is inferior in spread at the time of application.

10 Skin care cosmetics (sunscreen lotions)

Examples 1g to 7g, Comparative Example 1g

The respective materials of a component X shown in Tables 21 and 22 described hereinafter were blended in the proportion shown in the same tables and heated to 80 degree C to obtain the component X. The respective materials of a component Y were blended in the proportion shown in the same tables and heated to 80 degree C to obtain the component Y. Then, the component X and component Y were mixed at 80 degree C to prepare a sunscreen lotion, respectively.

Using the sunscreen lotions of the Examples and Comparative Example thus obtained, the respective characteristics were evaluated according to the criteria for skin care cosmetics. The results are summarized in Tables 21 and 22 described hereinafter.

Table 21  
(Sunscreen lotion)  
(Parts)

	Examples			
	1g	2g	3g	4g
Component X	Octyl cinnamate	7.50	7.50	7.50
	Octyl palmitate	5.00	5.00	5.00
	Cetyl alcohol	1.00	1.00	1.00
	Polyethylene glycol monostearate	1.50	1.50	1.50
	Poly(oxyethylene oxypropylene) methyl-polysiloxane copolymer	1.00	1.00	1.00
	Dimethylstearylamine	2.00	2.00	2.00
Component Y	Amphoteric urethane resin (a)	1.00	1.00	1.00
	Silicone polymer (Types)	0.50 (1)	0.50 (2)	0.50 (3) (4)
	Purified water	69.5	69.5	69.5
		5	5	5
	Triethanolamine (99%)	0.70	0.70	0.70
	Thickeners (2%)	10.0	10.0	10.0
		0	0	0
	Antiseptic	0.25	0.25	0.25
Feel				
Spread				

Table 22

(Sunscreen lotion) (Parts)

	Examples			Comparative Example
	5g	6g	7g	
Component X	Octyl cinnamate	7.50	7.50	7.50
	Octyl palmitate	5.00	5.00	5.00
	Cetyl alcohol	1.00	1.00	1.00
	Polyethylene glycol monostearate	1.50	1.50	1.50
	Poly(oxyethylene oxypropylene) methyl-polysiloxane copolymer	1.00	1.00	1.00
	Dimethylslearylamine	2.00	2.00	2.00
Component Y	Amphoteric urethane resin (a)	1.00	1.00	1.00
	Silicone polymer (Types)	0.50 (5)	0.50 (6)	0.50 (7)
	Purified water	69.5	69.5	70.0
		5	5	5
	Trethanolamine (99%)	0.70	0.70	0.70
	Thickeners (2%)	10.0	10.0	10.0
Feel	Antiseptic	0	0	0
		0.25	0.25	0.25
Spread				

As is apparent from the results shown in Tables 21 and 22, the sunscreen lotions of the Examples have very good feel and good spread at the time of application because the amphoteric urethane resin and silicone polymer are used in combination.

To the contrary, the sunscreen lotion of the Comparative Example has good feel because the amphoteric urethane resin is used, but is inferior in spread at the time of application.

#### 10 Make-up cosmetics (nail polishes)

##### Examples 1h to 7h, Comparative Example 1h

As shown in Tables 23 and 24 described hereinafter, a pigment was dispersed in deionized water in the proportion shown in the same tables and other components were added in the proportion shown in the same tables. The resultant was mixed until it was made homogenous with stirring and deaerated to prepare a nail polish, respectively.

Using the nail polishes of the Examples and Comparative Example thus obtained, the respective characteristics were evaluated according to the following criteria. The results are summarized in Tables 23 and 24 described hereinafter.



### Feel

The feel for skin care cosmetic was evaluated at a practical use test using ten panelists. Evaluation criteria are set as follows:

- 5                   : The number of persons, who did not feel tight and also felt soft, is 9 or more.
- : The number of persons, who did not feel tight and also felt soft, is within a range from 6 to 8.
- : The number of persons, who did not feel tight and also felt soft, is within a range from 2 to 5.
- 10               ×: The number of persons, who did not feel tight and also felt soft, is 1 or less.

### Spread

The spread for skin care cosmetic was evaluated at a practical use test using ten panelists. Evaluation criteria are set as follows:

- : The number of persons, who felt that the spread at the time of application is very good, is 9 or more.
- 20               : The number of persons, who felt that the spread at the time of application is very good, is within a range from 6 to 8.



Table 23

(Nail Polish) (Parts)

	Examples			
	1h	2h	3h	4h
Amphoteric urethane resin (a)	10.0	10.0	10.0	10.0
Silicone polymer (Types)	0.5 (1)	0.5 (2)	0.5 (3)	0.5 (4)
Deionized water	86.1	86.1	86.1	86.1
Bentonite	0.6	0.6	0.6	0.6
Pigment	2.5	2.5	2.5	2.5
Perfume	0.1	0.1	0.1	0.1
Antiseptic	0.1	0.1	0.1	0.1
Silicone defoamer	0.1	0.1	0.1	0.1
Feel				
Spread				

Table 24

(Nail polish) (Parts)

	Examples				Comparative Example
	5h	6h	7h	1h	
Water phase	Amphoteric urethane resin (a)				10.0
	0.5 (5)	0.5 (6)	0.5 (7)	—	—
	86.1	86.1	86.1	86.1	86.1
	0.6	0.6	0.6	0.6	0.6
Pigment	2.5	2.5	2.5	2.5	2.5
Perfume	0.1	0.1	0.1	0.1	0.1
Antiseptic	0.1	0.1	0.1	0.1	0.1
Silicone defoamer	0.1	0.1	0.1	0.1	0.1
Feel					
Spread					

As is apparent from the results shown in Tables 23 to 24, the nail polishes of the Examples have very good feel and good spread at the time of application because the amphoteric urethane resin and silicone polymer are used in combination.

To the contrary, the nail polish of the Comparative Example has good feel because the amphoteric urethane resin is used, but is inferior in spread at the time of application.

10 Make-up cosmetics (mascaras)

Examples 1i to 7i, Comparative Example 1i

As shown in Tables 25 and 26 described hereinafter, propylene glycol, triethanolamine, a viscosity modifier, and an antiseptic were blended with purified water in the proportion shown in the same tables and, after dissolving them at 80 degree C, the pigment was dispersed therein to form a water phase. Then, the respective materials of a component Z were blended in the proportion shown in the same tables and dissolved at 80 degree C to form an oil phase. The oil phase was added to the water phase and the mixture was emulsified by using a homogenizing mixer. Then, the respective materials of a component Y were gradually charged in the proportion shown in the same tables, followed by stirring

$$\frac{1}{\sqrt{2}} \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix}, \quad \frac{1}{\sqrt{2}} \begin{pmatrix} 1 & i \\ 0 & -i \end{pmatrix}, \quad \frac{1}{\sqrt{2}} \begin{pmatrix} 1 & -i \\ 0 & i \end{pmatrix}, \quad \frac{1}{\sqrt{2}} \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}$$

5

Table 25

(Mascara)		(Parts)			
		Examples			
		1i	2i	3i	4i
Component X	Propylene glycol	5.0	5.0	5.0	5.0
	Triethanolamine	1.0	1.0	1.0	1.0
	Thickener (2%)	10.0	10.0	10.0	10.0
	Antiseptic	0.5	0.5	0.5	0.5
	Pigment	10.0	10.0	10.0	10.0
	Purified water	51.0	51.0	51.0	51.0
Component Y	Amphoteric urethane resin (a)	10.0	10.0	10.0	10.0
	Silicone polymer	0.5	0.5	0.5	0.5
	(Types)	(1)	(2)	(3)	(4)
Component Z	Stearic acid	6.0	6.0	6.0	6.0
	Beeswax	6.0	6.0	6.0	6.0
Feel					
Spread					

Table 26

(Mascara)

(Parts)

	Examples			Comparative Example
	5i	6i	7i	
Component X	Propylene glycol	5.0	5.0	5.0
	Triethanolamine	1.0	1.0	1.0
	Thickener (2%)	10.0	10.0	10.0
	Antiseptic	0.5	0.5	0.5
	Pigment	10.0	10.0	10.0
	Purified water	51.0	51.0	51.0
Component Y	Ampholeric urethane resin (a)	10.0	10.0	10.0
	Silicone polymer	0.5	0.5	—
	(Types)	(5)	(6)	(7)
Component Z	Stearic acid	6.0	6.0	6.0
	Beeswax	6.0	6.0	6.0
Feel				
Spread				







Table 27

(Foundation) (Parts)

	Examples			
	1j	2j	3j	4j
Oil phase	Stearic acid	2.4	2.4	2.4
	Propylene glycol monostearate	2.0	2.0	2.0
	Cetostearyl alcohol	0.2	0.2	0.2
	Liquid lanolin	2.0	2.0	2.0
	Liquid paraffin	3.0	3.0	3.0
	Isopropyl myristate	8.5	8.5	8.5
Water phase	Propyl paraoxybenzoate	q.s.	q.s.	q.s.
	Amphoteric urethane resin (a)	0.5	0.5	0.5
	Silicone polymer (Types)	0.3 (1)	0.3 (2)	0.3 (3)
	Deionized water	63.3	63.3	63.3
	Sodium carboxymethylcellulose	0.2	0.2	0.2
	Bentonite	0.5	0.5	0.5
Pigment	Propylene glycol	4.0	4.0	4.0
	Triethanolamine	1.1	1.1	1.1
	Methyl paraoxybenzoate	q.s.	q.s.	q.s.
	Titanium oxide	8.0	8.0	8.0
	Talc	4.0	4.0	4.0
	Coloring pigment	q.s.	q.s.	q.s.
Perfume		q.s.	q.s.	q.s.
Feel				
Spread				

Table 28

(Foundation) (Parts)

	Examples			Comparative Example
	5j	6j	7j	
Oil phase	Stearic acid	2.4	2.4	2.4
	Propylene glycol monostearate	2.0	2.0	2.0
	Cetostearyl alcohol	0.2	0.2	0.2
	Liquid lanolin	2.0	2.0	2.0
	Liquid paraffin	3.0	3.0	3.0
	Isopropyl myristate	8.5	8.5	8.5
	Propyl paraoxybenzoate	q.s.	q.s.	q.s.
	Amphoteric urethane resin (a)	0.5	0.5	0.5
Water phase	Silicone polymer (Types)	0.3 (5)	0.3 (6)	0.3 (7)
	Deionized water	63.3	63.3	63.6
	Sodium carboxymethylcellulose	0.2	0.2	0.2
	Bentonite	0.5	0.5	0.5
	Propylene glycol	4.0	4.0	4.0
	Triethanolamine	1.1	1.1	1.1
	Methyl paraoxybenzoate	q.s.	q.s.	q.s.
	Titanium oxide	8.0	8.0	8.0
Pigment	Talc	4.0	4.0	4.0
	Coloring pigment	q.s.	q.s.	q.s.
Perfume	q.s.	q.s.	q.s.	q.s.
Feel				
Spread				



When using an aqueous solution or a water dispersion of a silicone polymer as the silicone polymer, the compatibility with the amphoteric urethane resin is enhanced to some degree, thereby making it possible to prepare a cosmetic having good stability.

When structural units derived from ethylene oxide, as a nonionic hydrophilic component, is introduced into the structure of the amphoteric urethane resin, sufficient hydrophilicity is obtained and the hair washability are particularly improved when using as the hair cosmetic.

When a polysiloxane bond(s) is/are introduced into the structure of the above amphoteric urethane resin, the touch is particularly improved furthermore when using as the hair cosmetic.

What is claimed is:

1. A cosmetic comprising an amphoteric urethane resin having carboxyl group(s) and tertiary amino group(s) in a molecule, and silicone polymer.

2. The cosmetic according to claim 1, wherein said silicone polymer is a nonionic silicone polymer.

3. The cosmetic according to claim 1, wherein said silicone polymer is a polyether-modified silicone polymer.

4. The cosmetic according to claim 1, wherein said silicone polymer is a phenyl-modified silicone polymer.

5. The cosmetic according to claim 1, wherein said silicone polymer is an amino-modified silicone polymer.

6. The cosmetic according to claim 1, wherein said silicone polymer is an alkyl-modified silicone polymer.

7. The cosmetic according to claim 1, wherein said silicone polymer is an alkoxy-modified silicone polymer.

5 8. The cosmetic according to claim 1, wherein said silicone polymer is a cyclic silicone polymer.

9. The cosmetic according to any one of claims 1 to 8, wherein said silicone polymer is in the form of an  
10 aqueous solution or a water dispersion.

10. The cosmetic according to any one of claims 1 to 9, wherein said amphoteric urethane resin has structural unit(s) derived from ethylene oxide in its  
15 structure.

11. The cosmetic according to any one of claims 1 to 9, wherein said amphoteric urethane resin has polysiloxane bond(s) in its structure.

20

12. The cosmetic according to any one of claims 1 to 11, wherein said amphoteric urethane resin is in the form of an aqueous solution.

25





[illegible]

5

**DECLARATION FOR PATENT APPLICATION**

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name.

I believe that I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled **COSMETICS** the specification of which

(check one) ☒ is attached hereto  
☐ was filed on \_\_\_\_\_ as  
 Application Serial No. \_\_\_\_\_  
 and was amended on \_\_\_\_\_ (if applicable).

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to the patentability of this application in accordance with Title 37, Code of Federal Regulations, §1.56(a).

I hereby claim foreign priority benefits under Title 35, United States Code, §119 of any foreign application(s) for patent or inventor's certificate listed below and have also identified below any foreign application for patent or inventor's certificate having a filing date before that of the application on which priority is claimed:

Prior Foreign Application(s)	PCT	10 August 1999	Priority Claimed
<u>PCT/US00/21874</u> (Number)	<u>PCT</u> (Country)	<u>10 August 1999</u> (Day/Month/Year Filed)	Yes No
<u>                    </u> (Number)	<u>                    </u> (Country)	<u>                    </u> (Day/Month/Year Filed)	Yes No
<u>                    </u> (Number)	<u>                    </u> (Country)	<u>                    </u> (Day/Month/Year Filed)	Yes No

I hereby claim the benefit under Title 35, United States Code, §120 of any United States application(s) listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States application in the manner provided by the first paragraph of Title 35, United States Code, §112, I acknowledge the duty to disclose material information as defined in Title 37, Code of Federal Regulations, §1.56(a), which occurred between the filing date of the prior application and the national or PCT international filing date of this application:

(Application Serial No.)	(Filing Date)	(Status-patented, pending, abandoned)
<u>                    </u>	<u>                    </u>	<u>                    </u>
<u>                    </u>	<u>                    </u>	<u>                    </u>

I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and to transact all business in the Patent and Trademark Office connected herewith:

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I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

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Declaration

Docket No. **1888P**

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